## DISPERSION OF MASS BY MOLECULAR AND TURBULENT DIFFUSION: ONE-DIMENSIONAL CASE\*

BY

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1. Introduction. If a solute is placed in a solvent in turbulent motion, it is dispersed both by molecular and turbulent diffusion. We derive here, in a one-dimensional case, a formula for the mean concentration of solute, as a function of x and t, in terms of its initial distribution, the coefficient of molecular diffusion, and the statistical characteristics of the turbulent velocity field.

The one-dimensional problem for the infinite domain is treated as follows. Using the initial condition expressed by Eq. (2) below, an initial value problem (for the concentration of solute) is formulated for the diffusion-convection differential equation, Eq. (1), which contains a rather arbitrary convection velocity v(x, t) which is a function of x and t. The solution is obtained in the form of a perturbation series, by perturbing with respect to the "magnitude" of the convection velocity, and the nth order term of this series involves, besides the initial data and the coefficient of diffusion, an n-fold product of convection velocity factors, each evaluated at a different point and time. This solution is found to be valid at small dispersion times or for small intensities of turbulence.

Now let the convection velocity be a random (though still continuous and sufficiently differentiable) function of x and t; that is, let it be a member of an ensemble of functions, where the ensemble represents the turbulent velocity field. (This is the approach taken in the mathematical theory of turbulence.) When an ensemble average is taken of each term of the previously obtained perturbation series, there results a power series representation for the mean concentration in which the nth order term contains the nth order correlation coefficient of the turbulent velocity field. This technique, of introducing random functions into the theory of partial differential equations, is not a new one. Kampé de Fériet, for instance, has treated several classical initial value problems with random initial data (see  $[1, 2, 3]^1$ ).

The initial value problem is solved in Sec. 2. In Sec. 3 a physical interpretation is given of a sufficient condition for uniform convergence of the perturbation series solution. In Sec. 4 turbulence is introduced, in the manner indicated above, and in Sec. 5 the following example is considered: dispersion at small times for initial distributions and space correlation coefficients of Gaussian type.

The investigation described in this paper is now being extended in two directions. On the one hand, the treatment presented here will be applied to the corresponding problems in two and three dimensions. On the other hand, the formal operations employed here, especially those involved in the probability approach to turbulence, must be justified.

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line of inquiry. Also, he wishes to thank his colleagues, O. J. Deters and R. J. Rubin, for several helpful suggestions.

2. Solution by perturbation of the initial value problem for the diffusion-convection differential equation. The one-dimensional equation governing molecular diffusion in the presence of a convection velocity is

$$\frac{\partial s}{\partial t} - D \frac{\partial^2 s}{\partial x^2} = -\frac{\partial (sv)}{\partial x} \tag{1}$$

(see [4], vol. II, p. 593), where s(x, t) = concentration (mass per unit length) of dispersing matter, D = (constant) coefficient of molecular diffusion, v(x, t) = convective velocity, assumed to depend on x and t in such a way that v,  $\partial v/\partial x$  and  $\partial^2 v/\partial x^2$  are continuous in x and t together.

For later purposes we assume further that v(x, t),  $v_x(x, t)$  and  $v_{xx}(x, t)$  are uniformly bounded in an infinite strip  $-\infty < x < \infty$ ,  $0 \le t \le t_0$ , i.e., that there are finite positive numbers  $v^m$ ,  $v^m_x$ , and  $v^m_{xx}$ , which are the least upper bounds of |v(x, t)|,  $|v_x(x, t)|$  and  $|v_{xx}(x, t)|$ , respectively, for  $-\infty < x < \infty$ ,  $0 \le t \le t_0$ .

Consider the initial value problem consisting of Eq. (1) and the initial condition

$$s(x, 0) = f(x) \qquad (-\infty < x < \infty) \tag{2}$$

in the domain  $-\infty < x < \infty$ ,  $0 \le t \le t_0$ . The function f(x) is a given non-negative-valued function of x, twice continuously differentiable and uniformly bounded for  $-\infty < \xi < \infty$ . We introduce the dimensionless quantities

$$\tau = t/T, \qquad \tau_0 = t_0/T, \qquad \xi = x/(TD)^{1/2}, \qquad \sigma(\xi, \tau) = (TD)^{1/2} s(x, t)/Q,$$

$$\phi(\xi) = (TD)^{1/2} f(x)/Q, \qquad \Omega = (T/D)^{1/2} V, \qquad \omega(\xi, \tau) = v(x, t)/V.$$
(3)

where T is some characteristic time for the diffusion process,  $Q = \int_{-\infty}^{\infty} f(x) dx$  is the total mass present in the system at t = 0, and V is some measure of the magnitude of the convective velocity. In the case of a specific (i.e., non-random) convection velocity v(x, t) we could let  $V = v^m$ , while in the turbulent case we shall let V equal the root mean square turbulent velocity.

In terms of these dimensionless quantities, the initial value problem becomes

$$\frac{\partial \sigma}{\partial \tau} - \frac{\partial^2 \sigma}{\partial \xi^2} = -\Omega \frac{\partial (\sigma \omega)}{\partial \xi} \qquad (-\infty < \xi < \infty, 0 \le \tau \le \tau_0)$$
 (4)

$$\sigma(\xi,0) = \phi(\xi) \qquad (-\infty < \xi < \infty). \tag{5}$$

It follows from Eq. (3) and the definition of Q that

$$\int_{-\infty}^{\infty} \phi(\xi) \ d\xi = 1. \tag{6}$$

Assuming that the solution of (4) is expressible as a power series in  $\Omega$ :

$$\sigma(\xi, \tau) = \sum_{i=0}^{\infty} \Omega^{i} \sigma_{i}(\xi, \tau), \qquad (7)$$

we find, after substituting this series for  $\sigma(\xi, \tau)$  in (4), collecting all terms on the left hand side of the equation, and setting the coefficient of each power of  $\Omega$  equal to zero, that

$$\frac{\partial \sigma_0}{\partial \tau} - \frac{\partial^2 \sigma_0}{\partial \xi^2} = 0, \tag{8}$$

$$\frac{\partial \sigma_i}{\partial \tau} - \frac{\partial^2 \sigma_i}{\partial \xi^2} = -\frac{\partial}{\partial \xi} (\omega \sigma_{i-1}) \qquad i = 1, 2, \cdots$$
 (9)

The solution to (8) satisfying the initial condition (5) is

$$\sigma_0(\xi, \tau) = \int_{-\infty}^{\infty} \phi(\xi') \Gamma(\xi - \xi', \tau) d\xi', \qquad (10)$$

where

$$\Gamma(\xi, \tau) = (4\pi\tau)^{-1/2} \exp(-\xi^2/4\tau)$$
 (11)

is the so-called fundamental solution of the heat-conduction equation (8). Furthermore,  $\sigma_0(\xi, \tau)$  satisfies the normalization condition

$$\int_{-\infty}^{\infty} \sigma_0(\xi, \tau) d\xi = 1. \tag{12}$$

It can be shown, by the use of Duhamel's theorem (see [6]) and an integration by parts, that when  $\omega(\xi, \tau)$  is uniformly bounded in  $-\infty < \xi < \infty$ ,  $0 \le \tau \le \tau_0$ , a solution of (9) which vanishes at  $\tau = 0$  is given by

$$\sigma_i(\xi, \tau) = -\int_0^{\tau} \frac{d\tau'}{[\pi(\tau - \tau')]^{1/2}} \int_{-\infty}^{\infty} \omega \sigma_{i-1} e^{-\alpha^2} \alpha \, d\alpha, \qquad (13)$$

where  $\omega$  and  $\sigma_{i-1}$  are evaluated at  $[\xi + 2(\tau - \tau')^{1/2}\alpha, \tau']$ . Thus when  $\sigma_0$  and  $\sigma_{i-1}(i = 1, 2, \cdots)$  are given by (10) and (13) respectively, the infinite series (7) solves the initial value problem consisting of Eqs. (4) and (5) (provided this series and those formed by differentiating it term by term converge uniformly in the infinite strip under consideration; this question will be discussed in the next section).

If in (13) we introduce the new variable of integration  $\xi' = \xi + 2(\tau - \tau')^{1/2}\alpha$ , we get

$$\sigma_{i}(\xi, \tau) = -\int_{0}^{\tau} d\tau' \int_{-\infty}^{\infty} \omega(\xi', \tau') \sigma_{i-1}(\xi', \tau') \frac{\partial}{\partial \xi} \Gamma(\xi - \xi', \tau - \tau') d\xi', \qquad (14)$$

where  $\Gamma(\xi, \tau)$  is given in (11);  $(\partial/\partial \xi)$  means differentiation with respect to the first argument). Then since  $\Gamma$  vanishes at  $\xi = \pm \infty$ , it can be shown, by inverting the order of integration, that  $\int_{-\infty}^{\infty} \sigma_i(\xi, \tau) d\xi = 0$  for  $i = 1, 2, 3, \cdots$ . This result, together with Eq. (12), implies that solution (7) conserves mass.

3. Sufficient condition for uniform convergence and its physical interpretation. It can be shown that series (7) and the series obtained by differentiating (7) term by term converge uniformly in the strip  $-\infty < \xi < \infty$ ,  $0 \le \tau \le \tau_0$  when

$$4\left(\frac{\tau_0}{\pi}\right)^{1/2}\Omega\left\{\lim_{\substack{-\infty<\xi,<\infty\\0\leq\tau'\leq\tau_0}}\left[\left|\omega(\xi',\,\tau')\right|+\left|\omega_{\xi}(\xi',\,\tau')\right|+\left|\omega_{\xi\xi}(\xi',\,\tau')\right|\right]\right\}<1. \tag{15}$$

Using Eqs. (3) to translate this condition into physical variables, we get  $4(t_0/D\pi)^{1/2}(v^m + v_x^m + v_{xz}^m) < 1$ , where  $v^m$ ,  $v_x^m$ , and  $v_{xz}^m$  are defined following Eq. (1).

This means that the power series is a valid representation of the solution of the initial value problem in the time interval  $0 \le t \le t_0$  either (1) when for  $t_0$  given arbitrarily, the bound on the convection velocity and its first two x-derivatives is sufficiently small, or (2) when for an arbitrary uniformly bounded convection field (twice continuously differentiable with respect to x),  $t_0$  is sufficiently small. In connection with case 2 (antici-

pating the next section by taking a turbulent velocity field as our convection field) it has been noted in [5], p. 99, in considering dispersion from a point source, that "the effects of molecular agitation on the dispersion are not always negligible as compared with the effects of turbulence; indeed, when the dispersion process starts, the former effect is greater than the latter."

4. Application to turbulence. Now let  $\omega(\xi, \tau)$  be a random function (though still possessing all the regularity properties with respect to  $\xi$  and  $\tau$  previously assumed for our convection velocity) with  $\langle \omega(\xi, \tau) \rangle = 0$  at every point  $(\xi, \tau)$ . The case of a uniform non-zero mean velocity,  $\langle \omega \rangle$ , can be reduced to the present case by introducing a new space variable,  $\eta = \xi - \langle \omega \rangle \tau$ . The angle brackets denote an ensemble or stochastic average. Thus we regard the convection (i.e., large-scale motion as compared with the molecular agitation) as arising from a turbulent velocity field with zero mean velocity. It is further assumed that  $\langle \omega^2(\xi, \tau) \rangle$  is a constant independent of  $\xi$  and  $\tau$ . For convenience we set  $\langle \omega^2 \rangle \equiv 1$ , which can be done by letting  $V = \langle v^2 \rangle^{1/2}$ , so that  $\Omega$  is now proportional to the root mean square turbulent velocity. (See the discussion following Eq. (3).)

Rather than  $\sigma$  itself, we are now interested in the *mean* concentration,  $\langle \sigma \rangle$ , which we propose to find by averaging each of the terms in the series (7). Since  $\sigma_0$ , given by (10), does not contain  $\omega$ , then  $\langle \sigma_0 \rangle = \sigma_0$ . From (14)

$$\sigma_{1}(\xi, \, \tau) \, = \, - \, \int_{0}^{\tau} \, d\tau' \, \int_{-\infty}^{\infty} \, \omega(\xi', \, \tau') \, \sigma_{0}(\xi', \, \tau') \, \frac{\partial}{\partial \xi} \, \Gamma(\xi \, - \, \xi', \, \tau \, - \, \tau') \, \, d\xi',$$

so that  $\sigma_1$  is linear in  $\omega$ , and since  $\langle \omega \rangle \equiv 0$ , it follows that

$$\langle \sigma_1(\xi, \tau) \rangle \equiv 0.$$
 (16)

Again, if we use (14) and the preceding expression for  $\sigma_1$ , we have

$$\begin{split} \sigma_2(\xi,\,\tau) \, &= \, \int_0^\tau \, d\tau' \, \int_{-\infty}^\infty \, \omega(\xi',\,\tau') \, \frac{\partial}{\partial \xi} \, \Gamma(\xi\,-\,\xi',\,\tau\,-\,\tau') \, \, d\xi' \\ &\qquad \qquad \times \, \int_0^{\tau'} \, d\tau'' \, \int_{-\infty}^\infty \, \omega(\xi'',\,\tau'') \sigma_0(\xi'',\,\tau'') \, \frac{\partial}{\partial \xi} \, \, \Gamma(\xi'\,-\,\xi'',\,\tau'\,-\,\tau'') \, \, d\xi''. \end{split}$$

Thus,  $\sigma_2$  involves a product of  $\omega$ 's evaluated at different times and points and therefore, when averaged, will contain the second-order space-time correlation coefficient of the turbulent velocity field. Introducing this correlation coefficient:

$$R_2(\xi_1 \ , \ \tau_1 \ ; \xi_2 \ , \ \tau_2) \ \equiv \ \frac{\langle \omega(\xi_1 \ , \ \tau_1)\omega(\xi_2 \ , \ \tau_2) \rangle}{\langle \omega^2 \rangle} \ = \ \langle \omega(\xi_1 \ , \ \tau_1)\omega(\xi_2 \ , \ \tau_2) \rangle,$$

we get from the expression above for  $\sigma_2$ ,

$$\langle \sigma_{2}(\xi, \tau) \rangle = \int_{0}^{\tau} d\tau' \int_{-\infty}^{\infty} \frac{\partial}{\partial \xi} \Gamma(\xi - \xi', \tau - \tau') d\xi' \int_{0}^{\tau'} d\tau''$$

$$\times \int_{-\infty}^{\infty} R_{2}(\xi', \tau'; \xi'', \tau'') \sigma_{0}(\xi'', \tau'') \frac{\partial}{\partial \xi} \Gamma(\xi' - \xi'', \tau' - \tau'') d\xi''.$$
(17)

Similarly, the expression for  $\sigma_n$  involves a product of n  $\omega$ 's, each with a different argument, and so  $\langle \sigma_n \rangle$  will involve the *n*th order correlation coefficient of the turbulent velocity field

$$R_n(\xi_1 , \tau_1 ; \xi_2 , \tau_2 ; \cdots ; \xi_n , \tau_n) \equiv \langle \omega(\xi_1 , \tau_1)\omega(\xi_2 , \tau_2) \cdots \omega(\xi_n , \tau_n) \rangle.$$

Thus, when the power series representing  $\langle \sigma \rangle$  is known to converge, a knowledge of all the correlation coefficients of the turbulent velocity field yields  $\langle \sigma \rangle$  exactly as a function of  $\xi$  and  $\tau$ , while if one knows the correlation coefficients  $R_n$  up through order n one has (in principle, assuming that all the integrations can be carried out) an nth order approximation to  $\langle \sigma \rangle$ .

5. Example: Dispersion at small times for initial distributions and space correlation coefficients of Gaussian type. We have seen that one situation in which the perturbation series (7) actually solves the initial value problem is when, for a given convection field (or class of convection fields), the time interval  $0 \le \tau \le \tau_0$  is sufficiently small. Let us, then, calculate expressions for  $\sigma_0(\xi, \tau)$  and  $\langle \sigma_2(\xi, \tau) \rangle$  at small values of  $\tau$ , for certain initial distributions and correlation functions. (We recall that the first-order term, $\langle \sigma_1 \rangle$ , vanishes.) In terms of the computation the restriction to small values of  $\tau$  is imposed by the desire to approximate certain functions of  $\tau$  so that they can be integrated (see Eq. (22) below).

Consider initial distributions of the form

$$\phi(\xi) = (4\pi\tau^*)^{-1/2} \exp(-\xi^2/4\tau^*),$$

depending on the parameter  $\tau^*$ . Then from (10)

$$\sigma_0(\xi, \tau) = [4\pi(\tau + \tau^*)]^{-1/2} \exp[-\xi^2/4(\tau + \tau^*)]. \tag{18}$$

(This is the distribution that would result from molecular diffusion of a unit mass concentrated at  $\xi = 0$ , starting at time  $\tau = -\tau^*$ .)

We assume that the turbulence is homogeneous and stationary, and furthermore that the second-order correlation coefficient can be written as a function of the  $\xi$ 's times a function of the  $\tau$ 's:

$$R_2(\xi',\,\tau';\,\xi'',\,\tau'') \,=\, \Re_2(\xi'\,-\,\xi'';\,\tau'\,-\,\tau'') \,=\, \Re_\xi(\xi'\,-\,\xi'') \Re_\tau(\tau'\,-\,\tau'').$$

Finally we consider the particular class of functions

$$\mathfrak{R}_{\xi}(\xi) = \exp\left(-\xi^2/4b\right),\tag{19}$$

with the parameter b. Without specifying  $\mathfrak{R}_{\tau}$ , we assume that in the neighborhood of  $\tau=0$  its rate of decrease is sufficiently slow so that for the small values of  $\tau$  under consideration  $\mathfrak{R}_{\tau}(\tau)\approx 1$  (since  $\mathfrak{R}_{\tau}(0)=1$ ). We shall, however, carry along the factor  $\mathfrak{R}_{\tau}$  in the calculations until it is necessary to invoke this assumption, so that perhaps some enterprising reader can find an explicit functional form for  $\mathfrak{R}_{\tau}$  which will obviate the approximations which are made at that point.

Under the foregoing assumptions, if we replace  $\xi''$  and  $\tau''$  by the new variables of integration  $\xi_2 = \xi' - \xi''$  and  $\tau_2 = \tau' - \tau''$ , Eq. (17) can be written

$$\langle \sigma_{2}(\xi, \tau) \rangle = -\frac{1}{4\pi^{1/2}} \int_{0}^{\tau} \frac{d\tau'}{(\tau - \tau')^{3/2}} \int_{-\infty}^{\infty} (\xi' - \xi) \exp\left[ -\frac{(\xi' - \xi)^{2}}{4(\tau - \tau')} \right] d\xi'$$

$$\times \frac{1}{4\pi^{1/2}} \int_{0}^{\tau'} \frac{\Re_{\tau}(\tau_{2}) d\tau_{2}}{\tau_{2}^{3/2}} \int_{-\infty}^{\infty} \xi_{2} \Re_{\xi}(\xi_{2}) \sigma_{0}(\xi' - \xi_{2}, \tau - \tau_{2}) \exp\left( -\frac{\xi_{2}^{2}}{4\tau_{2}} \right) d\xi_{2},$$
(20)

where  $\partial \Gamma/\partial \xi$  has been obtained from (11). Inserting the expressions for  $\sigma_0$  and  $\mathfrak{R}_{\xi}$  given by (18) and (19) into the first integral to be evaluated,

$$I_1^{\xi}(\xi', \tau', \tau_2) \equiv \int_{-\infty}^{\infty} \xi_2 \Re_{\xi}(\xi_2) \sigma_0(\xi' - \xi_2, \tau' - \tau_2) \exp\left(-\frac{\xi_2^2}{4\tau_2}\right) d\xi_2$$
,

we find, after an essentially simple integration, that

$$I_1^{\xi} = \frac{\xi' b^{3/2} \tau_2^{3/2}}{D^{3/2}} \exp{\left[-\frac{\xi'^2 (\tau_2 + b)}{4D}\right]},$$

where

$$D = \tau_2(\tau' - \tau_2 + \tau^*) + b(\tau' + \tau^*). \tag{21}$$

Next, we must evaluate

$$I^{\tau}(\xi', \tau') \equiv \frac{1}{4\pi^{1/2}} \int_{0}^{\tau'} \frac{\mathfrak{R}_{\tau}(\tau_{2})}{\tau_{2}^{3/2}} I_{1}^{\xi}(\xi', \tau', \tau_{2}) d\tau_{2}$$

$$= \frac{\xi' b^{3/2}}{4\pi^{1/2}} \int_{0}^{\tau'} \frac{\mathfrak{R}_{\tau}(\tau_{2})}{D^{3/2}} \exp\left[-\frac{\xi'^{2}(\tau_{2} + b)}{4D}\right] d\tau_{2} , \qquad (22)$$

where D is given by (21). Since  $\tau_2 \leq \tau' \leq \tau \ll 1$ ,  $\mathfrak{R}_{\tau}(\tau_2) \approx 1$ . Then, by considering different relative magnitudes for  $\tau$ ,  $\tau^*$  and b, we can simplify D and evaluate I' in three cases: (1)  $\tau \ll b$ ,  $\tau^* \ll b$ ; (2)  $\tau \ll b$ ,  $\tau \ll \tau^*$ ; (3)  $\tau \ll \tau^*$ ,  $b \ll \tau^*$ . In each case we shall evaluate  $I'(\xi', \tau')$ , then (see Eq. (20))

$$I_{2}^{\xi}(\xi, \, \tau, \, \tau') \equiv \int_{-\infty}^{\infty} (\xi' - \xi) \, \exp\left[-\frac{(\xi' - \xi)^{2}}{4(\tau - \tau')}\right] I^{\tau}(\xi', \, \tau') \, d\xi', \tag{23}$$

and finally

$$\langle \sigma_2(\xi, \tau) \rangle \equiv -\frac{1}{4\pi^{1/2}} \int_0^{\tau} \frac{I_2^{\xi}(\xi, \tau, \tau')}{(\tau - \tau')^{3/2}} d\tau'.$$
 (24)

Case 1:  $\tau \ll b$ ,  $\tau^* \ll b$ . From (21), neglecting terms of second order in the small quantities  $\tau$ ,  $\tau'$ ,  $\tau_2$  and  $\tau^*$ , we get  $D = b(\tau + \tau^*)$ . We insert this expression for D and  $\Re_{\tau} = 1$  in (22) to obtain

$$I^{\tau} = \frac{\xi'}{4\pi^{1/2}} \int_{0}^{\tau'} \exp\left[-\frac{\xi'^{2}}{4(\tau' + \tau^{*})}\right] \frac{d\tau_{2}}{(\tau' + \tau^{*})^{3/2}}$$
$$= \frac{\xi'\tau'}{4\pi^{1/2}(\tau' + \tau^{*})^{3/2}} \exp\left[-\frac{\xi'^{2}}{4(\tau' + \tau^{*})}\right].$$

Then, using this result in (23) and integrating gives

$$I_{2}^{\xi} = \frac{\tau'}{4\pi^{1/2}(\tau' + \tau^{*})^{3/2}} \int_{-\infty}^{\infty} \xi'(\xi' - \xi) \exp\left[-\frac{(\xi' - \xi)^{2}}{4(\tau - \tau')} - \frac{{\xi'}^{2}}{4(\tau' + \tau^{*})}\right] d\xi'$$
$$= -\frac{\tau'}{2} \frac{(\tau - \tau')^{3/2}}{(\tau + \tau^{*})^{5/2}} \left[\xi^{2} - 2(\tau + \tau^{*})\right] \exp\left[-\frac{\xi^{2}}{4(\tau + \tau^{*})}\right],$$

and at last from (24) we get

$$\langle \sigma_2(\xi, \tau) \rangle = \frac{\tau^2}{16\pi^{1/2}} \frac{[\xi^2 - 2(\tau + \tau^*)]}{(\tau + \tau^*)^{5/2}} \exp\left[-\frac{\xi^2}{4(\tau + \tau^*)}\right].$$
 (25)

Case 2:  $\tau \ll b$ ,  $\tau \ll \tau^*$ . Now  $D = b\tau^*$  and

$$I^{\tau} = \frac{\xi'}{4\pi^{1/2}\tau^{*3/2}} \int_{0}^{\tau'} \exp\left(-\frac{\xi'^{2}}{4\tau^{*}}\right) d\tau_{2} \ .$$

Proceeding as in Case 1, we find

$$\langle \sigma_2(\xi, \tau) \rangle = \frac{\tau^2}{16\pi^{1/2}} \frac{(\xi^2 - 2\tau^*)}{\tau^{*5/2}} \exp\left(-\frac{\xi^2}{4\tau^*}\right).$$
 (26)

Case 3:  $\tau \ll \tau^*$ ,  $b \ll \tau^*$ . Now  $D = \tau^*(\tau_2 + b)$ . By performing the successive integrations indicated above (and using along the way the fact that  $\tau' \ll \tau \ll \tau^*$ ), we obtain in this case

$$\langle \sigma_2(\xi, \tau) \rangle = \frac{\xi^2 - 2\tau^*}{4\pi^{1/2}\tau^{*5/2}} \exp\left(-\frac{\xi^2}{4\tau^*}\right) \left\{ b\tau - 2b^{3/2} [(\tau + b)^{1/2} - b^{1/2}] \right\}. \tag{27}$$

Remarks. 1. The expressions for  $\langle \sigma_2 \rangle$  in Cases 1 and 2, given by Eqs. (25) and (26) respectively, are quite similar in form; indeed, if in Case 1 we made the more restrictive assumption that  $\tau \ll \tau^* \ll b$ , (25) would reduce to (26). Equation (27), the result for Case 3, however, is different from the other two: only in this case does b appear in the expression for  $\langle \sigma_2 \rangle$ . Thus  $\langle A_2 \rangle = \sigma_0 + \Omega^2 \langle \sigma_2 \rangle$ , the second-order approximation to  $\langle \sigma_2 \rangle$ , does not depend on b when  $\tau \ll b$ , whereas in the absence of this assumption the expression for  $\langle A_2 \rangle$  does involve b. (It should be noted that b is proportional to  $L^2$ , where L is the length scale of turbulence, the integral of the correlation coefficient from 0 to  $\infty$  with respect to x.)

2. If in Case 3 the more restrictive assumption that  $\tau \ll b \ll \tau^*$  is made, we get  $\langle \sigma_2 \rangle = 0$ , for

$$(\tau + b)^{1/2} - b^{1/2} = b^{1/2} \left[ \left( 1 + \frac{\tau}{b} \right)^{1/2} - 1 \right] \approx b^{1/2} \left[ \left( 1 - \frac{\tau}{2b} \right) - 1 \right] = \frac{\tau}{2b^{1/2}}.$$

- 3. A comparison of Eqs. (18) and (25) shows that in Case  $2 \langle \sigma_2 \rangle = (\tau^2/2) \partial \sigma_0 / \partial \tau$ . It is not clear what significance, if any, there is in this interesting relation.
- 4. Within the limitations of requirement (15), our sufficient condition for the validity of the perturbation scheme, it can be shown that in the region of the  $\xi$ ,  $\tau$ -plane where

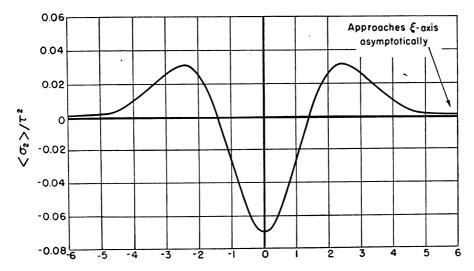


Fig. 1. Example of second-order correction to mean concentration at small times:  $\langle \sigma_2 \rangle / \tau^2$  plotted as a function of  $\xi$  from Eq. (26) with  $\tau^* = 1$ .

 $\langle A_2 \rangle$  is a good approximation to  $\langle \sigma \rangle$ ,  $\Omega^2 \langle \sigma_2 \rangle$  is much smaller than  $\sigma_0$ . (This is not surprising since in perturbing with respect to the convection term, we assumed this term is small in comparison with the others in the original differential equation.) It should be kept in mind, of course, that sufficient conditions for a certain result flow out of the particular mathematical technique employed, and often the result is valid under much broader conditions. This is particularly true of perturbation methods. Nevertheless, even if we limit ourselves to values of the parameters and variables which satisfy (15), the results are of interest. Equations (25), (26) and (27) show that in all three cases, for given values of b and  $\tau^*$  and a fixed value of  $\tau$ ,  $\langle \sigma_2 \rangle$  is negative at  $\xi^2 = 0$ , increases, with increasing  $\xi^2$ , to a positive maximum, and then approaches 0 asymptotically as  $\xi^2 \to \infty$ . This is illustrated in the figure for Case 2. The exponential factors in both  $\sigma_0$  and  $\langle \sigma_2 \rangle$ make them go to 0 as  $\xi^2 \to \infty$ , but the *relative* magnitude of  $\langle \sigma_2 \rangle$ , as compared with  $\sigma_0$ , increases with  $\xi^2$ ; indeed,  $\langle \sigma_2 \rangle / \sigma_0$  becomes proportional to  $\xi^2$  as  $\xi^2$  increases. Adding turbulence to molecular diffusion, then, "sweeps out" the initial distribution more rapidly, and this additional dispersive effect assumes a particular functional form, at least in this set of examples.

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